

allowance of the claim are respectfully requested in light of the following remarks.

The Examiner has rejected claim 1 under 35 U.S.C. § 102(b) for purported anticipation by, or in the alternative, under 35 U.S.C. § 103(a) for purported obviousness over the teachings of United States Patent No. 5,322,888 to Kato et al. (Kato). The Examiner asserts that Kato discloses composite sols of Optolake 1130F-2 which is a  $\text{SiO}_2/\text{TiO}_2/\text{Fe}_2\text{O}_3$  composite having a particle size of 20 nm in methanol, that this coating composition is surface modified with  $\gamma$ -glycidoxypropyltriethoxysilane, and that the dielectric constant for methanol at 20°C is 31.2. The Examiner further asserts that the  $\gamma$ -glycidoxypropyltriethoxysilane disclosed by Kato has a molecular polarizability in the range claimed in the present application because of its close structural similarity to  $\gamma$ -glycidoxypropyltrimethoxysilane and  $\gamma$ -glycidoxypropylmethoxydiethoxysilane, which the present application teaches as examples of compounds having the claimed molecular polarizability. Alternatively, the Examiner asserts that Kato teaches both  $\gamma$ -glycidoxypropyltriethoxysilane and  $\gamma$ -glycidoxypropyltrimethoxysilane as treating agents and makes no distinction between them, so that it would have been obvious to substitute the latter for the former as an obvious functional equivalent.

The Examiner has rejected claim 1 under 35 U.S.C. § 102(e) for purported anticipation by or, in the alternative, under 35 U.S.C. § 103(a) for purported obviousness over the teachings of U.S. Patent No. 5,858,077 to Kayanoki (Kayanoki '077). The Examiner asserts that Kayanoki '077 discloses compositions comprising a composite metal oxide dispersed in methanol and/or isopropanol with the further addition of a polymerizing monomer. The Examiner also asserts that Kayanoki '077 teaches treating the methanol dispersed composite particle sol with  $\gamma$ -glycidoxypropyltrimethoxysilane followed by the addition of isopropanol and a polymerization monomer. The Examiner then asserts that

Kayanoki '077 discloses a dispersion medium dielectric constant equivalent to that of the present invention and that  $\gamma$ -glycidoxypropyltrimethoxysilane has the molecular dispersibility claimed in the present application. The Examiner also asserts that, although Kayanoki '077 claims the further addition of a polymerization monomer, there is no evidence showing that the dielectric constant differs as a result, or that the present invention is rendered nonobvious thereby. The Examiner also asserts that U.S. Patent No. 5,654,090 to Kayanoki (Kayanoki '090) is cumulative to the rejection over Kato et al. and Kayanoki '077.

Applicants maintain that the present invention can be distinguished from the Kato, Kayanoki '077 and Kayanoki '090 patents.

#### The Present Invention

Particles of inorganic oxides must generally have their surfaces rendered hydrophobic for obtaining an organic solvent dispersion thereof. Thus, generally, the surfaces of such particulates are modified.

A method of modifying the surface of such inorganic compound particulates comprises reacting, for example, a reactive monomer or a coupling agent with hydroxyl groups of the particulate surface. However, it is difficult to obtain an organic solvent sol of high dispersibility using this method.

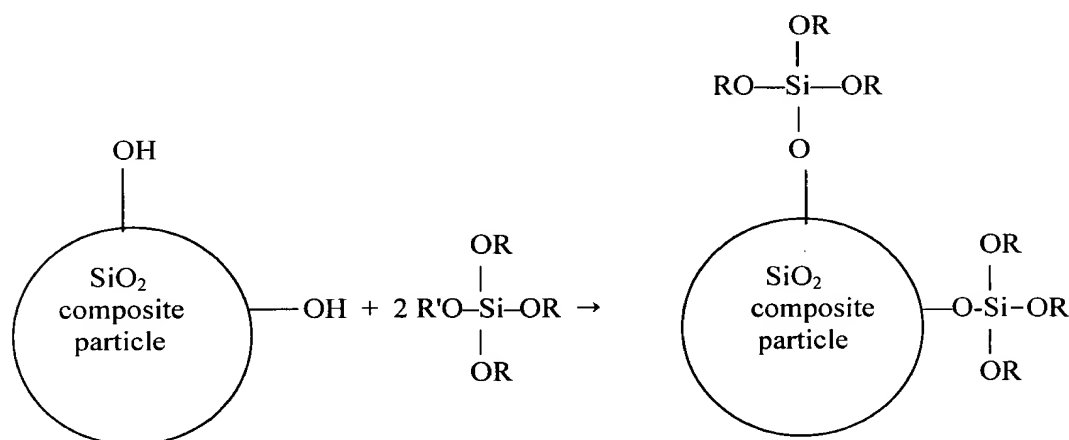
When the above inorganic compound particles are used as a filler in, for example, a coating material, a hard coating agent component of an insulating coat or a protective coat, adding a sol thereof to a matrix of coating film forming agent is likely to produce an aggregation of particulates in the matrix. In particular, a problem has been encountered in that, when the organic solvent sol is used in the presence of a cation, anion, or surfactant, particulates may be aggregated and a gelation may occur. For example, in order to improve the hardness, water resistance and stain resistance of coating films in coating materials, an organic solvent sol in which the inorganic compound has been dispersed has

been used. However, a problem has been encountered in that the coating material becomes viscous and whitens because the organic compound sol has low compatibility with the coating film formed resin and the resin emulsion being produced.

These problems have been solved in the present invention.

In the present invention, SiO<sub>2</sub> composite particles are modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds,  $\gamma$ -mercaptopropyltrimethoxysilane and  $\gamma$ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ , wherein the SiO<sub>2</sub> composite particles are composite oxide particulates composed of silica and at least one inorganic oxide other than silica.

Modification by organosilane is caused by condensation of -OH groups on the SiO<sub>2</sub> composite particle surface and organosilane as shown by the following drawing.



In the organic compound sol of the present invention, the surface of the SiO<sub>2</sub> composite particles is modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, and epoxysilane organic compounds having specified molecular polarizability. Therefore, the SiO<sub>2</sub> composite particles have a desirable affinity with the

dispersion media, so that the dispersion stability thereof in the dispersion media is excellent without the occurrence of aggregation of particulates and gelation. Further, even if an organic or inorganic acid or a salt thereof were present in the inorganic compound sol, the SiO<sub>2</sub> composite particles would not aggregate with each other and gelation would not occur.

When the inorganic compound sol of the present invention is blended as a filler in, for example, a coating material or a hard coating agent, even if an acid, base, salt or surfactant is present in the inorganic compound sol, the resulting coating film is substantially free from transparency decrease and cracking attributed to the aggregation of particulates and gelation. Therefore, the inorganic compound sol of the present invention is useful as a filler to be blended in not only various coating materials and hard coating agents but also various resins.

U.S. Patent No. 5,322,888

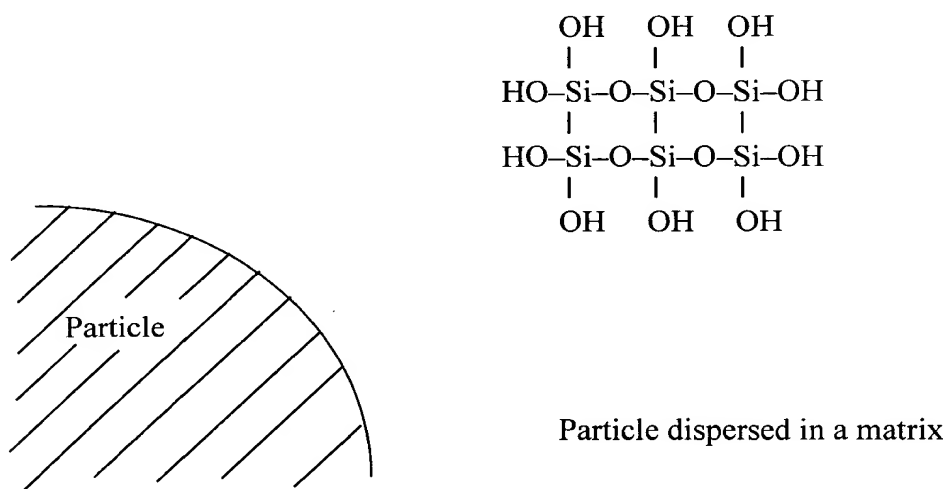
U.S. Patent No. 5,322,888 (Kato) contains a description of a coating composition for optical plastic moldings comprising a hydrolyzate of a silane compound, titanium oxide-based composite fine particles, an unsaturated or saturated polycarboxylic acid or an anhydride thereof, and a heat-curing catalyst. (See claim 1)

(1) The particles used in the present invention differ from the particles used by Kato. In the present invention, the SiO<sub>2</sub> composite particles are composite oxide particles composed of silica and at least one oxide other than silica. In contrast, Kato uses titanium oxide-based composite fine particles in which titanium oxide combines integrally with iron oxide. Kato never teaches SiO<sub>2</sub> composite particles.

(2) In the present invention, the SiO<sub>2</sub> composite particles are modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds,  $\gamma$ -mercaptopropyltrimethoxysilane and  $\gamma$ -

chloropropyltrimethoxysilane exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40}$   $\text{C}^2\text{m}^2\text{J}^{-1}$ . In contrast, Kato teaches the use of a hydrolyzate of a silane compound as a coating forming component (matrix), not for modifying the titanium oxide-based composite particles (see column 3, lines 46-57).

If the hydrolyzate of a silane compound is mixed with titanium oxide-based composite fine particles according to the teachings of Kato, the resulting components are titanium oxide-based composite fine particles dispersed in a hydrolyzate of a silane compound (matrix) as shown in the following illustration:



If the hydrolyzate of a silane compound is mixed with fine particles of a titanium oxide-based composite, the titanium oxide-based composite fine particles are not modified by the hydrolyzate of a silane compound.

(3) Kato states that the titanium oxide-based composite fine particles may be treated with a silane coupling agent for surface modification. Tetramethoxysilane, methyltrimethoxysilane, trimethylchlorosilane, vinyltriethoxysilane,  $\gamma$ -glycidoxypropyltriethoxysilane, and  $\gamma$ -glycidoxypropylmethyldiethoxysilane are given as examples of the silane coupling agent (see column 5, lines 26 to 44 of Kato). However, the selective use of specific organic compounds selected from vinylsilane compounds,

acrylsilane compounds, epoxysilane compounds, aminosilane compounds,  $\gamma$ -mercaptopropyltrimethoxysilane and  $\gamma$ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40} \text{ C}^2\text{m}^2\text{J}^{-1}$  as a silane coupling agent is never described by Kato.

The modification of the surface of the  $\text{SiO}_2$  composite particles by an organosilane selected from vinylsilane compounds, acrylsilane compounds, or epoxysilane, and having specified molecular polarizability causes the  $\text{SiO}_2$  composite particles to have the desired affinity with the dispersion media, so that the dispersion stability thereof in the dispersion media are excellent, and aggregation of particulates and gelation do not occur. Further, even if an organic or inorganic acid or a salt thereof were present in the inorganic compound sol, the  $\text{SiO}_2$  composite particles would not aggregate with each other and gelation would not occur. This effect is made clear by the comparison of Example 1 and Comparative Example 2:

**Table 1**

	Sol Composition	Modifying organic compound		Dispersion Medium	Stability	
					Sulfuric acid	Hydrochloric acid
Example 1	$\text{SiO}_2 \cdot \text{Al}_2\text{O}_3$	$\gamma$ -glycidoxypentyltrimethoxysilane	$10.4 \times 10^{-40}$	Ethylene glycol	Stable at least 6 months	Stable at least 6 months
Comp. Example 2	$\text{SiO}_2 \cdot \text{Al}_2\text{O}_3$	Monomethyltrimethoxysilane	$1.9 \times 10^{-40}$	Ethylene glycol	Gelled 30 days	Gelled 40 days

As shown in Table 1, when the modifying organic compound has a molecular polarizability higher than  $2 \times 10^{-40} \text{ C}^2\text{m}^2\text{J}^{-1}$ , the resulting organic compound sol has excellent dispersibility in organic solvent even if inorganic acids are present in the sol. (See Example 1)

In contrast, when the modifying organic compound has a molecular polarizability of lower than  $2 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ , the resulting organic compound sol has poor dispersibility in organic solvent. (See Comparative Example 2)

This effect is not suggested by Kato, which does not disclose the selective use of specific organic compounds exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ .

As was mentioned above, the coating composition of Kato differs from that of the present invention. Therefore, the effect of particles modified according to the present invention is neither disclosed nor suggested by Kato.

U.S. Patent No. 5,564,090

Kayanoki '090 describes a coating composition comprising (1) particles of at least one oxide selected from the group consisting of silica, iron oxide, titanium oxide, cerium oxide, zirconium oxide, antimony oxide, zinc oxide and tin oxide, and composites thereof, (2) an epoxy group-containing silicon compound or a partial hydrolyzate thereof, (3) an organic compound having the formula  $\text{R}^4\text{-X-R}^5\text{YH}$ , wherein  $\text{R}^4$  represents a monovalent hydrocarbon group having at least one unsaturated group,  $\text{R}^5$  represents a divalent hydrocarbon group having two or more carbon atoms and contains an oxygen atom or a sulfur atom, X represents  $\text{-O-}$ ,  $\text{-CO-O-}$ ,  $\text{-S-}$ ,  $\text{-CO-S-}$  and  $\text{-CS-S-}$ , and Y represents an oxygen atom or a sulfur atom, and (4) a curing catalyst for said epoxy group-containing silicon compound. Kayanoki '077 describes coating compositions in a manner differing from that of Kayanoki '090 only in that fewer oxides are claimed, and in that the formula of organic compound (3) is expressed in a slightly different manner.

Kayanoki '090 teaches the use of an epoxy group-containing silicon compound or a partial hydrolyzate thereof as a coating forming component (matrix) and not for modifying the oxide particles. Therefore, the amount of an epoxy group-containing silicon compound or partial hydrolyzate thereof in the coating composition of Kayanoki '090 is much greater than the amount of organosilane compound used in the present invention. In the present invention, the amount of organosilane compound used is very small. For example, 8.4 g of organosilane compound are used in combination with 90 g of SiO<sub>2</sub> composite particles (9.3% by weight) in Example 1 of the present invention. In contrast, according to Kayanoki '090 and Kayanoki '077, the epoxy group-containing silicon compound or a partial hydrolyzate thereof is used in an amount of from 5 to 90 wt.%, and the fine particles are used in an amount from 10 to 70 wt.%, based on the total solids in the coating composition. That is, Kayanoki '090 and Kayanoki '077 teach the use of a weight of the epoxy group-containing silicon compound or a partial hydrolyzate that is at least 50% of the weight of the fine particles.

Kayanoki '090 and Kayanoki '077 teach that the fine particles may be subjected to surface modification with organosilicon compounds. A wide range of organosilicon compounds is suggested (see column 3, line 16 to column 4, line 16 of Kayanoki '090, and column 3, line 12 to column 4, line 18 of Kayanoki '077). However, Kayanoki '090 and Kayanoki '077 do not teach the selective use of specific organic compounds selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds,  $\gamma$ -mercaptopropyltrimethoxysilane and  $\gamma$ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$  as a silane coupling agent.



As mentioned above, modification of the surface of SiO<sub>2</sub> composite particles by an organosilane, having specified molecular polarizability, selected from vinylsilane compounds, acrylsilane compounds, or epoxysilane compounds produces SiO<sub>2</sub> composite particles having desirable affinity with the dispersion media, so that the stability thereof in the dispersion media is excellent without the occurrence of aggregation of particulates or gelation. Further, even if an organic or inorganic acid or salt thereof were present in the inorganic compound sol, the SiO<sub>2</sub> composite particles would not aggregate with each other and the gelation would not occur.

Such an effect is not suggested by Kayanoki '090 and Kayanoki '077, which do not disclose the selective use of specific organic compounds exhibiting a molecular polarizability of  $2 \times 10^{-40}$  to  $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ . Therefore, the effect of a particulate produced according to the present invention is neither disclosed nor suggested by Kayanoki '090 or Kayanoki '077.

In view of the above, it is submitted that the claim is in condition for allowance. Reconsideration of the rejections is requested. Allowance of claim 1 at an early date is solicited.

Respectfully submitted,

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